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Collective charge density fluctuations in superconducting layered systems with bilayer unit cells

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Collective modes of bilayered superconducting superlattices (e.g., YBCO) are investigated within the conserving gauge-invariant ladder diagram approximation including both the nearest interlayer single electron tunneling and the Josephson-type Cooper pair tunneling. By calculating the density-density response function including Coulomb and pairing interactions, we examine the two collective mode branches corresponding to the in-phase and out-of-phase charge fluctuations between the two layers in the unit cell. The out-of-phase collective mode develops a long wavelength plasmon gap whose magnitude depends on the tunneling strength with the mode dispersions being insensitive to the specific tunneling mechanism (i.e., single electron or Josephson). We also show that in the presence of tunneling the oscillator strength of the out-of-phase mode overwhelms that of the in-phase-mode at $k_\parallel = 0$ and finite k_z, where k_z and $k_\parallel$ are respectively the mode wave vectors perpendicular and along the layer. We discuss the possible experimental observability of the phase fluctuation modes in the context of our theoretical results for the mode dispersion and spectral weight.

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I. INTRODUCTION

In contrast to bulk isotropic superconductors, where longitudinal collective modes (i.e., plasmons) associated with the density fluctuation response is virtually of no particular interest or significance in the context of superconducting properties, there has been substantial recent theoretical interest in the longitudinal collective mode spectra of layered high-T_c superconductors[1–6]. This interest arises primarily from the highly anisotropic two dimensional layered structure of these materials which, in principle, allow for sub-gap plasmon modes residing inside the superconducting gap in the low wave vector regime. This gives rise to interesting collective mode behavior[1–10] in layered anisotropic superconductors which have no analogs in bulk isotropic superconductors. In this paper we consider the effect of having multilayer complex unit cells, as existing in YBCO and BISCO high-T_c superconductor materials, on the longitudinal electronic collective mode spectrum. We find a number of collective modes arising from the complex unit cell structure, and comment on their possible experimental relevance. One of our goals is to critically assess whether observable electronic collective mode behavior could shed some light on the interesting and unusual mechanism producing high T_c superconductivity in these materials. The other goal is to predict novel collective mode behavior peculiar to layered superconductors with no analogs in bulk systems.

The collective mode spectrum is characterized by the energy dispersion ($\hbar = 1$ throughout this paper) relation $\omega \equiv \omega(k_\parallel, k_z)$, which we calculate in this paper, where $k_\parallel \equiv |\mathbf{k}_\parallel|$ is the two dimensional wave vector in the so-called a-b plane (along the layer) and k_z is the wave vector along the c-axis, then $k_z = |\mathbf{k}|\cos\theta$, $k_\parallel = |\mathbf{k}|\sin\theta$. Because of the strong a-b plane versus c-axis anisotropy in these materials, the dependence of the collective mode frequency on $k_\parallel$ and k_z is very different. [We ignore any anisotropy, which is invariably rather small, in the a-b plane and assume intralayer planar isotropy, i.e., $\omega(\mathbf{k}_\parallel, k_z) \equiv \omega(k_\parallel, k_z)$.] The structural model we employ considers the layered superconductor to be a one dimensional superlattice along the z direction (c-axis) composed of a periodic system of bilayer unit cells with an intracell layer separation of c and a superlattice period of d ($> c$). The two active layers separated by a distance c within each unit cell are taken to be identical and are assumed to be planar two dimensional electron gas (2D EG) systems of charge density n_s per unit area and zero layer thickness each. In most of our calculations presented in this paper the intercell electron hopping (or tunneling) between neighboring unit cells (separated by a distance d) is neglected (i.e., we neglect any superlattice band width along the z direction), but we critically examine the effect of intracell electron hopping between the two layers within each unit cell on the collective mode dispersion. We comment upon the effect of a finite *inter*cell hopping in the conclusion of this article. We include in our theory the long range (intracell and intercell) Coulomb interaction among all the layers. This long range Coulomb interaction, which couples all the layers, is of great importance in determining

the collective mode spectrum. We also include in our theory of collective mode dispersion the effect of the superconducting pairing interaction, assumed in our model to be a short-range in-plane attractive interaction of the BCS-Fermi liquid type, which is treated in a fully gauge invariant Nambu-Gorkov formalism. Our work is thus a generalization of the earlier work[1,2] by Fertig and Das Sarma, and by Hwang and Das Sarma (who considered only the monolayer superconducting superlattice situation with only a single layer per unit cell) to a complex unit cell situation with two layers per unit cell. To keep the situation simple we will consider only the s-wave gap symmetry[1], which, according to ref. 2 gives a very good account of the collective mode dispersion even for the d-wave case except at very large wave vectors. Following the work of Fertig and Das Sarma[1] there has been a great deal of theoretical and experimental work[2–10] on the electronic collective mode properties in layered superconducting materials, but the specific issue considered in this paper has not earlier been discussed in the literature for a multilayer superconducting system. It should also be pointed out that, while the focus of our work is the collective mode behavior in layered high-T_c cuprate superconductors (which are *intrinsic* superlattice systems due to their highly anisotropic crystal structure with CuO layers), our results equally well describe *artificial* superconducting superlattices made of multilayer metallic structures provided $k_\parallel$ and k_z are good wave vectors in the system.

The collective mode dispersion in bilayered superconducting superlattices is quite complicated. There are essentially two different branches of long wavelength collective modes: in-phase (ω_+) modes and out-of-phase (ω_-) modes, depending on whether the electron density fluctuations in the two layers are in-phase or out-of-phase. Each of these collective modes disperses as a function of wave vector, showing strong anisotropy in $k_\parallel$ and k_z dispersion. In particular, the limits ($k_z = 0$, $k_\parallel \to 0$) and ($k_z \to 0$, $k_\parallel = 0$) are *not* equivalent because the $k_z = 0$ three dimensional limit is singular. For $k_z = 0$ the in-phase ω_+ collective mode is a gapped three dimensional plasma mode at long wavelengths ($k_z = 0$, $k_\parallel \to 0$) by virtue of the Higgs mechanism arising from the long range Coulomb interaction coupling all the layers. This mode characterizes the long wavelength in-phase charge fluctuations of all the layers. For non-zero k_z the ω_+ mode vanishes at long wavelengths ($k_\parallel \to 0$) because at finite k_z the system is essentially two dimensional. The out-of-phase ω_- collective mode branch arises purely from the bilayer character of the system and indicates the out-of-phase density fluctuations in the two layers. In the absence of any interlayer hopping (either intracell and intercell) the ω_- mode is purely acoustic in nature vanishing at long wavelengths ($k_\parallel \to 0$) as $\omega_-(k_z, k_\parallel \to 0) \sim O(k_\parallel)$ independent of the value of k_z. For finite interlayer tunneling ω_- exhibits a tunneling gap at $k_\parallel = 0$. The Higgs gap for $\omega_+(k_z = 0, k_\parallel \to 0)$ is not qualitatively affected by intracell interlayer tunneling because the three dimensional plasma energy is usually substantially larger then the tunneling energy.

Note that, in the absence of any intracell and intercell tunneling, both in-phase and out-of-phase collective mode branches lie below the superconducting energy gap for small $k_\parallel$ [except for the $\omega_+(k_z = 0)$ mode which is pushed up to the three dimensional plasma frequency]. This remains true even for weak intracell and intercell tunnelings, and in this paper we concentrate mainly on this long wavelength "below gap" regime where the phase fluctuation modes could possibly lie in the superconducting gap. For simplicity we also restrict ourselves to s-wave gap symmetry of the superconducting order parameter. This approximation may at first sight appear to be unusually restrictive as it seems to rule out the applicability of our theory to bilayer high-T_c materials (such as YBCO, BISCO) which are now widely accepted to have d-wave ground state symmetry. This, however, is not the case because at long wavelengths (small $k_\parallel$), which is what we mostly concentrate on, the collective mode spectrum is insensitive to the order parameter symmetry[2], and therefore our results apply equally well to high-T_c bilayer materials. The modes we predict and their dispersion should most easily be observable via the resonant inelastic light scattering spectroscopy, but may also be studied via frequency domain far infrared spectroscopy using a grating coupler.

II. THEORY, APPROXIMATIONS, AND RESULTS

In our calculation we assume that the two layers in each unit cell can be considered to be 2D EG, and all layers are identical, having the same 2D charge density n_s per unit area. Two identical layers separated by a distance c in each unit cell are strongly coupled through the interlayer intracell electron tunneling. The interlayer tunneling is between the well-defined CuO layers in high T_c materials. The intercell tunneling between different unit cells separated by a distance d (in our model $d > c$) is neglected at first (see section III for the effect of intercell tunneling). Although we neglect the electron tunneling between different unit cells, the electrons in *all* layers are coupled via the intercell long range Coulomb potential which we keep in our theory. Since the long wavelength plasma modes are independent of the gap function symmetry[2], we work in the BCS approximation with s-wave pairing for simplicity. Then, in the Nambu representation[11] the effective Hamiltonian of a bilayered superconductor with 2D quasiparticle energy $\varepsilon(k)$, a tight-binding coherent single-electron intracell hopping $t(k)$, and an additional coherent intracell Josephson coupling T_J between two nearest layers is given by

$$H = H_0 - \mu N + H_{\rm int} + H_{T_J}, \tag{1}$$

with

$$H_0 - \mu N = \sum_{n,i} \sum_{\mathbf{k}} \tilde{\varepsilon}_{\mathbf{k}} \Psi^{\dagger}_{\mathbf{k},ni} \tau_3 \Psi_{\mathbf{k},ni} + \sum_{n,i} \sum_{\mathbf{k}} t(\mathbf{k}) \Psi^{\dagger}_{\mathbf{k},ni} \tau_3 \Psi_{\mathbf{k},n\bar{i}}, \tag{2}$$

$$H_{\rm int} = \frac{1}{2} \sum_{ni,mj} \sum_{\mathbf{q}} \rho_{\mathbf{q},mi} \tilde{V}_{mi,nj}(\mathbf{q}) \rho_{-\mathbf{q},nj}, \tag{3}$$

$$H_{T_J} = \sum_{n,i} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} T_J \left(\Psi_{\mathbf{k}+\mathbf{q},ni} \tau_3 \Psi_{\mathbf{k},n\bar{i}} \right) \left(\Psi_{\mathbf{k}'-\mathbf{q},ni} \tau_3 \Psi_{\mathbf{k}',n\bar{i}} \right), \tag{4}$$

where n, m are the unit cell indices and i, $j = 1, 2$ label the two layers within a given unit cell ($\bar{i} = 3 - i$). Here, $\Psi_{\mathbf{k},ni}$ and $\Psi^{\dagger}_{\mathbf{k},ni}$ are the column and row vectors

$$\Psi_{\mathbf{k},ni} \equiv \begin{pmatrix} c_{\mathbf{k},ni,\uparrow} \\ c^{\dagger}_{-\mathbf{k},ni,\downarrow} \end{pmatrix}, \qquad \Psi^{\dagger}_{\mathbf{k},ni} \equiv \left(c^{\dagger}_{\mathbf{k},ni,\uparrow}, c_{-\mathbf{k},ni,\downarrow} \right), \tag{5}$$

where $c^{\dagger}_{\mathbf{k},ni,\sigma}$ ($c_{\mathbf{k},ni,\sigma}$) creates (annihilates) an electron with wave vector $\mathbf{k}$ and spin σ in the i-th layer within the nth unit cell, and $\rho_{\mathbf{q},ni}$ denotes the density operator defined by

$$\rho_{\mathbf{q},ni} = \sum_{\mathbf{k}} \Psi^{\dagger}_{\mathbf{k}+\mathbf{q},ni} \tau_3 \Psi_{\mathbf{k},ni}, \tag{6}$$

where $\tilde{\varepsilon}_{\mathbf{k}} = k^2/2m - \mu$ (μ being the chemical potential), and τ_i (i=1,2,3) are the Pauli matrices. In Eq. (3), the effective interaction $\tilde{V}_{ni,mj}(\mathbf{q})$ contains the long range Coulomb interaction coupling all the layers and the short range attractive intralayer pairing interaction (giving rise to superconductivity in the problem)

$$\tilde{V}_{ni,mj}(\mathbf{q}) = V_c(q_{\|}) \exp[-q_{\|}|z_{ni} - z_{mj}|] + V_0 \delta_{n,m} \delta_{i,j} \tag{7}$$

where $V_c(q_{\|}) = 2\pi e^2/(\kappa q_{\|})$ is the two dimensional Coulomb interaction and κ is the background dielectric constant of the system. V_0 represents a weak, short-ranged attractive intra-layer pairing interaction which produces superconductivity, and is a model parameter in our theory.

We should comment on one unusual feature of our Hamiltonian defined in Eqs. (1)–(4). This is the existence of *both* a coherent single-particle hopping term, defined by the single-particle hopping amplitude $t(k)$ in Eq. (2), and a coherent Cooper pair Josephson tunneling term, defined by T_J in Eq. (4). Usually the existence of a single-particle hopping t automatically generates an equivalent Josephson coupling T_J in the superconducting system, and keeping both of them as we do, namely, t in the single particle Hamiltonian H_0 [Eq. (2)] and T_J in the two-particle Josephson coupling [Eq. (4)], is redundant. We do, however, wish to investigate separately effects of both coherent single particle hopping and Josephson coupling along the c-axis on the collective mode spectra because of recent suggestions[12] of a novel interlayer tunneling mechanism for superconductivity in cuprates which explicitly postulates $t = 0$ (in the normal state) and $T_J \neq 0$ (in the superconducting state). Our model therefore uncritically includes both t and T_J as distinct contributions, and one could think of the interlayer Josephson coupling T_J in our model Hamiltonian arising from some interlayer pairing interaction not included in our model pairing interaction V_0 which is exclusively intralayer in nature. In the following we take t and T_J to be independent parameters of our model without worrying about their microscopic origins.

The collective modes of the system are given by the poles of the reducible density response function $\chi(\mathbf{k}, \omega)$. We apply the conserving gauge invariant ladder diagram approximation[1,11] in calculating the density response of the system including the effect of the pairing interaction induced vertex and self-energy corrections. The density response function is defined as

$$\chi(\mathbf{k}, \omega) = -i \int_0^{\infty} dt e^{i\omega t} < [\rho(\mathbf{k}, t), \rho(-\mathbf{k}, 0)] >, \tag{8}$$

where $\rho(\mathbf{k}, t)$ is the three dimensional Fourier transform of the density operator in the Heisenberg representation. Here, $\mathbf{k} \equiv (k_{\|}, k_z)$ is the 3D wave vector, where k_z measures the wave vector along the z-axis (i.e., the c-direction) and $k_{\|}$ is the 2D x-y plane (i.e., a-b plane) wave vector. The density response may be written in terms of an irreducible polarizability $\Pi(\mathbf{k}, \omega)$ as shown in Fig. 1(a). Following Anderson's arguments for the absence of single particle tunneling[12] we first neglect inter-layer single electron tunneling effects ($t = 0$) and only consider the

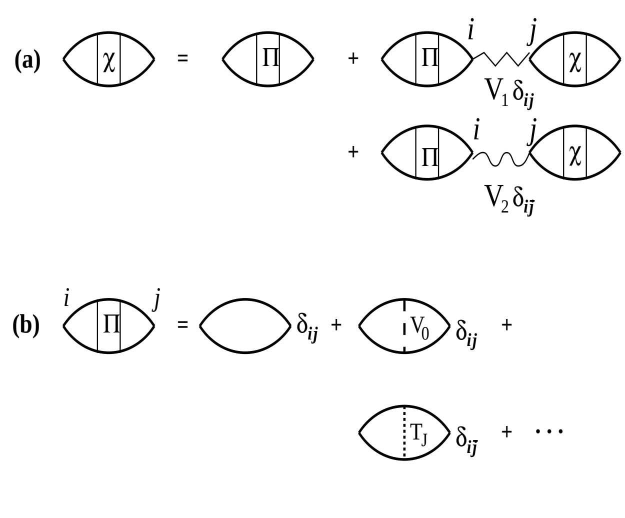

FIG. 1. (a) Diagrammatic representation of the dielectric response in terms of the irreducible polarizability Π. Here, V_1 and V_2 are given in Eq. (13), and $\bar{j} = 3 - j$. (b) Irreducible polarizability used in this calculation.

Josephson coupling effect. The polarizability Π is then diagonal in the unit cell index and becomes the corresponding 2D polarizability matrix, $\Pi(\mathbf{k},\omega) \equiv \Pi(k_\|,\omega)$

$$\chi(\mathbf{k},\omega) = \frac{\Pi(k_\|,\omega)}{\epsilon(\mathbf{k},\omega)}, \tag{9}$$

where $\Pi(k_\|,\omega)$ is the irreducible polarizability matrix for a single isolated unit cell,

$$\Pi(k_\|,\omega) = \begin{pmatrix} \Pi_{11}(k_\|,\omega) & \Pi_{12}(k_\|,\omega) \\ \Pi_{21}(k_\|,\omega) & \Pi_{22}(k_\|,\omega) \end{pmatrix}, \tag{10}$$

where Π_{11}, Π_{22} and Π_{12}, Π_{21} indicate the intra-layer and inter-layer irreducible polarizability, respectively. Within our approximation, the inter-layer polarizabilities vanish when the single-particle tunneling is neglected. We will see that the plasma gap of the out-of-phase mode arises entirely from the non-vanishing inter-layer irreducible polarizability. In Eq. (9) the effective dynamical dielectric function $\epsilon(\mathbf{k},\omega)$ is given by

$$\epsilon(\mathbf{k},\omega) = \mathbf{1} - \tilde{V}(\mathbf{k})\Pi(k_\|,\omega), \tag{11}$$

where $\mathbf{1}$ is a 2×2 unit matrix and $\tilde{V}(\mathbf{k})$ is the effective interaction which in our simple model is given by

$$\tilde{V}(\mathbf{k}) = \begin{pmatrix} V_1(\mathbf{k}) & V_2(\mathbf{k}) \\ V_2(\mathbf{k}) & V_1(\mathbf{k}) \end{pmatrix}, \tag{12}$$

where $V_1(\mathbf{k})$ corresponds to the intralayer interaction ($i=j$) and $V_2(\mathbf{k})$ the interlayer interaction ($i\neq j$), which arises entirely from the long-range Coulomb coupling in our model, and they are given by

$$\begin{aligned} V_1(\mathbf{k}) &= V_c(k_\|)f(\mathbf{k}) + V_0, \\ V_2(\mathbf{k}) &= V_c(k_\|)g(\mathbf{k}), \end{aligned} \tag{13}$$

where $f(\mathbf{k})$ and $g(\mathbf{k})$, the superlattice form factors which modify the 2D Coulomb interaction due to Coulomb coupling between all the layers in our multilayer superlattice system, are given by

$$f(\mathbf{k}) = \frac{\sinh(k_\| d)}{\cosh(k_\| d) - \cos(k_z d)}, \tag{14}$$

$$g(\mathbf{k}) = \frac{\sinh[k_\|(d-c)] + e^{-ik_z d}\sinh(k_\| c)}{\cosh(k_\| d) - \cos(k_z d)} e^{ik_z c}. \tag{15}$$

In order to obtain the collective mode spectrum, it is necessary to construct a gauge invariant and number-conserving approximation for $\Pi(\mathbf{k},\omega)$. In the conserving gauge invariant ladder diagram approximation[1,11] the irreducible polarizability obeys the ladder integral equation which is shown diagrammatically in Fig. 1(b). It may be written in the form

$$\begin{aligned} \Pi_{i,j}(k,\omega) = -i\mathrm{Tr}\int \frac{d\omega_1 dp_1}{(2\pi)^3}\tau_3 G_i(p_1,\omega_1) \\ \times\Gamma_{i,j}(p_1,k,\omega)G_i(k-p_1,\omega-\omega_1), \end{aligned} \tag{16}$$

where $G_i(k,\omega)$ is the i-th layer Green's function with self-energy corrections (self-consistent Hartree approximation in the Coulomb interaction and self-consistent Hatree-Fock approximation in the short-range pairing interaction) and $\Gamma_{i,j}$ is a vertex function. The vertex part satisfies the linear integral equation

$$\begin{aligned} \Gamma_{ij}\ (\ p_1,k,\omega) = \tau_3\delta_{ij} + i\sum_{l=1}^{2}\int \frac{d^2q d\omega_1}{(2\pi)^3}\tau_3 G_l(q,\omega_1) \\ \times\Gamma_{ij}(q,k,\omega)G_l(q-k_1,\omega-\omega_1)\tau_3\left[V_0\delta_{li} + T_J\delta_{\bar{l}i}\right], \end{aligned} \tag{17}$$

where $\bar{l} = 3-l$. In order to solve this vertex function, we expand Γ_{ij} in Pauli matrices

$$\Gamma_{ij} = \sum_{l=0}^{3}\gamma_{ij,l}\tau_l. \tag{18}$$

Since our model assumes two identical 2D layers in the unit cell, we have $\Gamma_{11} = \Gamma_{22} = \Gamma_a$ and $\Gamma_{12} = \Gamma_{21} = \Gamma_b$. By introducing the polarization function

$$\begin{aligned} P_i &= i\int \frac{d^2q d\omega_1}{(2\pi)^3}\tau_3 G(q,\omega)\tau_i G(q-k,\omega_1-\omega)\tau_3 \\ &= \sum_{j=0}^{3}\bar{P}_{i,j}\tau_j, \end{aligned} \tag{19}$$

the vertex function, Eq. (17), becomes

$$\begin{pmatrix} \gamma_a \\ \gamma_b \end{pmatrix} = \begin{pmatrix} \mathbf{I}_3 \\ 0 \end{pmatrix} + V_0\begin{pmatrix} \underline{P}\gamma_a \\ \underline{P}\gamma_b \end{pmatrix} + T_J\begin{pmatrix} \underline{P}\gamma_b \\ \underline{P}\gamma_a \end{pmatrix}, \tag{20}$$

where γ's are column vectors, $I_3^T = (0,0,0,1)$, and $\underline{P}$ is a 4×4 matrix whose components are given by $\bar{P}_{ij}$ in Eq. (19). Then, the polarizability function Π_{ij} becomes

$$\begin{aligned} \Pi_{ij} &= -\mathrm{Tr}\sum_{l=0}^{3}\bar{P}_{i,l}\tau_3\gamma_{ij,l} \\ &= -\sum_{l=0}^{3}\left[P_i\gamma_{ij}\right]_{3,l}. \end{aligned} \tag{21}$$

The poles of the vertex function or polarizability Π give the collective mode spectra for the neutral superconductor (i.e., neglecting the long range Coulomb coupling which couples all the layers). In the long wavelength limit we have two collective modes ("phasons") for the *neutral* system

$$\omega_+^2(k) = (v_0 k)^2\left[1 + (V_0+T_J)N_0/2\right], \tag{22}$$

$$\omega_-^2(k) = \omega_0^2 + v_0^2 k^2\left[1 + N_0(V_0-T_J)/2\right], \tag{23}$$

where $v_0 = v_F/\sqrt{2}$ with v_F as the Fermi velocity, $N_0 = m/\pi$ is the 2D density of states at the Fermi surface, and $\omega_0^2 = 16T_J\Delta^2/[N_0(V_0^2 - T_J^2)]$ is the tunneling gap induced by the finite Josephson coupling

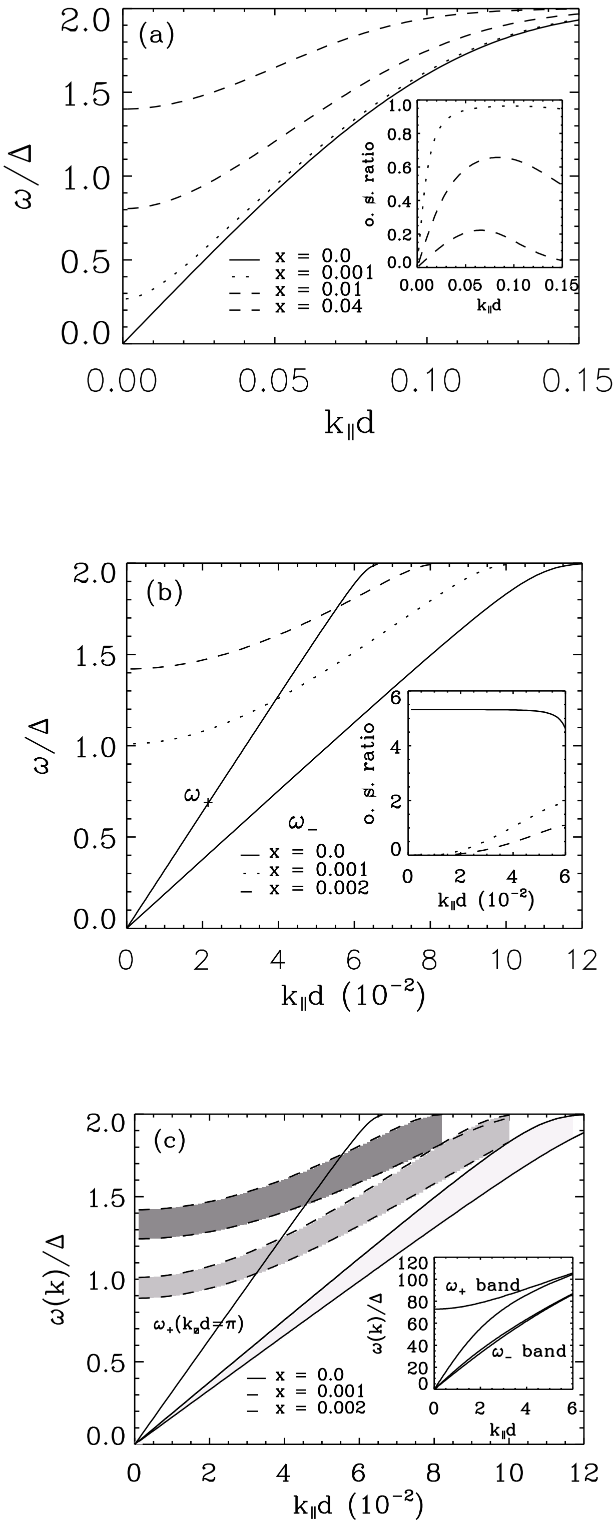

FIG. 2. (a) The plasmon mode ($\omega_\pm$) dispersions in the presence of Josephson tunneling for the neutral bilayered superconducting superlattice as a function of $k_\parallel$ for fixed $k_z d = \pi$. Here, $x = T_J/V_0$ indicates the Josephson tunneling strength with respect to the intra-layer pairing interaction. Inset shows the ratio of the oscillator strength of ω_+ to that of ω_-. (b) The plasmon mode dispersions ($\omega_\pm$) for the charged system. Inset shows the ratio of the oscillator strength of ω_- to that of ω_+. (c) The $\omega_-(\mathbf{k})$ band in the superlattice for the charged system as a function of in-plane wave vector ($k_\parallel d$) in the presence of the tunneling. Inset shows the $\omega_\pm$ band of the bilayer superconducting superlattice. We use parameters roughly corresponding to YBCO in these figures: the sheet density $n_s = 10^{14} cm^{-2}$, effective in-plane mass $m = 5m_0$, lattice dielectric constant $\kappa = 4$, $d = 12\mathring{A}$, and $c = 3\mathring{A}$.

($T_J \neq 0$). The ω_+ mode corresponds to the in-phase motion of the order parameter, or, equivalently the 2-D Goldstone-Anderson-Bogoliubov phase fluctuation mode due to the spontaneously broken continuous gauge symmetry of the superconducting state. The ω_- mode corresponds to the out-of-phase mode first predicted for a two-band superconductor[13], which has recently been calculated within the time-dependent Hartree-Fork-Gor'kov (mean-field) approximation[4] for a two-layer superconductor system. In Fig. 2(a) we show the calculated collective mode dispersion for different Josephson tunneling strengths with respect to the intra-layer pairing interaction, $x = T_J/V_0$. When the Josephson tunneling is absent, $x = 0$, the two phason modes $\omega_\pm$ are degenerate and have identical dispersion (solid line). But in the presence of finite Josephson tunneling between the nearest layers, $x \neq 0$, the out-of phase mode (ω_-) develops a plasma gap (ω_0) depending on the tunneling strength.The in-phase mode ω_+ is not affected qualitatively by finite Josephson tunneling and remains an acoustic Goldstone mode (i.e., $\omega_+ \sim O(k)$ for $k \to 0$) although the velocity of the acoustic plasmon does depend on T_J (cf. Eq. (22)). In Fig. 2(a) the inset shows the relative oscillator strength of the two phason modes, the ratio of the spectral weight of ω_- to that of ω_+. The ratio decreases as tunneling amplitude increases. This is due to the approach of the ω_- mode to the pair-breaking excitation region ($\omega \approx 2\Delta$) at large tunneling, which causes decay of the ω_- mode to single particle excitations, and the strength of the mode transfers to pair-breaking excitations. These results apply to any bilayered *neutral* superconductors (which, of course, do not exist in nature because Coulomb interaction is always present in real systems).

By looking for zeros of the dynamical dielectric function defined by Eq. (11) we find the collective modes of the charged superconducting superlattices. Since the two layers within the cell are identical we have $\Pi_{11} = \Pi_{22}$ and $\Pi_{12} = \Pi_{21}$, which gives rise to distinct in-phase and out-of-phase collective charge density fluctuations of the charged superconductor. Coupling of the in-phase (out-of-phase) mode of the neutral system via the long range Coulomb interaction to the charge density fluctuation of

the layers gives rise to the in-phase (out-of-phase) collective mode of the charged bilayer system. The dielectric function is a matrix, and the zeros of the det$[\epsilon]$, which define the collective mode spectra, are given by

$$\begin{aligned}\det[\epsilon] &= [1-(\Pi_{11}+\Pi_{12})(V_1+V_2)] \\ &\quad\times[1-(\Pi_{11}-\Pi_{12})(V_1-V_2)] = 0. \qquad (24)\end{aligned}$$

In the long wavelength limit Eq. (24) can be analytically solved using Eqs. (13) – (21), and we find two distinct collective modes corresponding to the relative phase of the charge density fluctuations in the two layers within each unit cell:

$$\omega_+^2(\mathbf{k}) = \omega_p^2 \frac{k_\parallel d}{4}\left[f(\mathbf{k}) + |g(\mathbf{k})|\right]_{k_\parallel\to 0}, \qquad (25)$$

$$\omega_-^2(\mathbf{k}) = \frac{(1+\Delta V - \Delta V_0)(\omega_0^2 + v_0^2 k^2/2)}{1-\omega_0^2(\Delta V - \Delta V_0)/6}, \qquad (26)$$

where $\omega_p = (4\pi n_B e^2/\kappa m)^{1/2}$ is a three dimensional plasma frequency with the effective three-dimensional electron density of the double-layered supperlattice $n_B = 2n_s/d$, and $k^2 = k_\parallel^2 + k_z^2$ with $\mathbf{k} \equiv (k_\parallel, k_z)$; $\Delta V = N_0(V_1 - V_2)$ and $\Delta V_0 = N_0(V_0 - T_J)/2$. In Fig. 2(b) we show the calculated charge density mode dispersion for fixed $k_z d = \pi$ as a function of $k_\parallel d$. Tunneling has little effect on the in-phase mode (thin solid line) but profoundly affects the out-of-phase mode (thick lines) by introducing a gap at $\omega_-(k_\parallel = 0)$ similar to the neutral case. Since in the presence the tunneling the out-of-phase mode acquires a gap, the two modes cross at the resonant frequency ($\omega_+ = \omega_-$), but the symmetry ("parity") associated with the two identical layers does not allow any mode coupling or anti-crossing effect. If the two layers in the unit cell are not identical then there is a mode coupling induced anti-crossing around $\omega_+ \approx \omega_-$. The inset shows the ratio of the oscillator strength of the in-phase mode to that of the out-of-phase mode. In sharp contrast to the neutral system, in the long wavelength limit the out-of-phase mode ω_- completely dominates the spectral weight in the presence of interlayer tunneling. In the absence of tunneling ($x = 0$), however, the in-phase mode ω_+ dominates the spectral weight. Our results for the collective mode dispersion in the presence of finite single-particle tunneling but vanishing Josephson coupling (i.e., $t \neq 0$, $T_J = 0$) are qualitatively identical to the situation with $t = 0$, $T_J \neq 0$, and are therefore not shown separately. This is, of course, the expected result because t automatically generates an effective Josephson tunneling, i.e., an effective T_J, in the superconducting system, and therefore the qualitative effect of having a finite T_J or a finite t in the superconducting system is similar.

We also calculate the collective modes of the bilayered superconducting system by including *both* the single particle tunneling and the Josephson tunneling between the nearest layers (i.e., $t, T_J \neq 0$). The two layers in the unit cell hybridized by the single particle tunneling matrix element, $t(\mathrm{k})$, would lead to a symmetric and an antisymmetric combination of the quasiparticle states for each value of the wave vector $\mathbf{k}$ in the plane. By introducing the symmetric and antisymmetric single electron operators with respect to an interchanging of the two layers, $\alpha_{n,k,\sigma} = \frac{1}{\sqrt{2}}(c_{n1,k\sigma} + c_{n2,k\sigma})$ and $\beta_{n,k,\sigma} = \frac{1}{\sqrt{2}}(c_{n1,k\sigma} - c_{n2,k\sigma})$, the total effective Hamiltonian can be written as

$$\begin{aligned}H &= \sum_n \sum_{k\sigma}\left[\alpha^\dagger_{n,k\sigma}\varepsilon_1(k)\alpha_{n,k\sigma} + \beta^\dagger_{n,k\sigma}\varepsilon_2(k)\beta_{n,k\sigma}\right] \\ &+ \frac{1}{2}\sum_{nn'}\sum_{\mathbf{q}}\left[\rho^T_{1,n\mathbf{q}}\bar{U}(\mathbf{q})\rho_{1,n'-\mathbf{q}} + \rho^T_{2,n\mathbf{q}}\bar{V}(\mathbf{q})\rho_{2,n'-\mathbf{q}}\right], \quad (27)\end{aligned}$$

where $\varepsilon_1(k) = \varepsilon(k) + t(k)$ and $\varepsilon_2(k) = \varepsilon(k) - t(k)$, and

$$\rho_{1,n\mathbf{q}} = \sum_{\mathbf{k}\sigma}\begin{pmatrix}\alpha^\dagger_{n,\mathbf{k+q}\sigma}\alpha_{n,\mathbf{k}\sigma} \\ \beta^\dagger_{n,\mathbf{k+q}\sigma}\beta_{n,\mathbf{k}\sigma}\end{pmatrix}, \qquad (28)$$

$$\rho_{2,n\mathbf{q}} = \sum_{\mathbf{k}\sigma}\begin{pmatrix}\alpha^\dagger_{n,\mathbf{k+q}\sigma}\beta_{n,\mathbf{k}\sigma} \\ \beta^\dagger_{n,\mathbf{k+q}\sigma}\alpha_{n,\mathbf{k}\sigma}\end{pmatrix}, \qquad (29)$$

and

$$\bar{U}(\mathbf{q}) = \begin{pmatrix} U_+ & U_- \\ U_- & U_+\end{pmatrix}, \quad \bar{V}(\mathbf{q}) = \begin{pmatrix} V_+ & V_- \\ V_- & V_+\end{pmatrix}, \qquad (30)$$

where $U_\pm = V_1 + V_2 \pm T_J$ and $V_\pm = V_1 - V_2 \pm T_J$. This Hamiltonian is identical to that in the corresponding two subband model, which is well studied in semiconductor quantum well systems[14]. Since there are no off-diagonal elements of the interaction with respect to the subband index we have well separated intra-subband and inter-subband collective modes corresponding to the in-phase and out-of-phase modes, respectively. Within our gauge-invariant ladder diagram approximation we can easily calculate the mode dispersions by following the procedure outlined in Eqs. (5) – (26). The in-phase-mode for both the neutral and the charged system is insensitive to tunneling in the long wavelength limit, and has essentially the same long wavelength dispersion as in Eq. (22) and Eq. (25) respectively, up to second order in k. The out-of-phase mode is, however, strongly affected by both the coherent single particle tunneling and the Josephson tunneling, and has a dispersion

$$\omega_-^2(k) = \omega_0^2 + \left[(2t)^2 + v_0^2 k^2\right]\left[1 + \Delta V_0\right], \qquad (31)$$

for neutral superconductors, and

$$\omega_-^2(\mathbf{k}) = \frac{(1+\Delta V - \Delta V_0)\left[\omega_0^2 + (2t)^2 + v_0^2 k_\parallel^2\right]}{1 - \frac{\omega_0^2}{6}(\Delta V - \Delta V_0)}, \qquad (32)$$

for charged systems in the presence of finite tunneling. In Fig. 3, we show the calculated mode dispersions as a function of the in-plane wave vector $k_\parallel d$ for a fixed

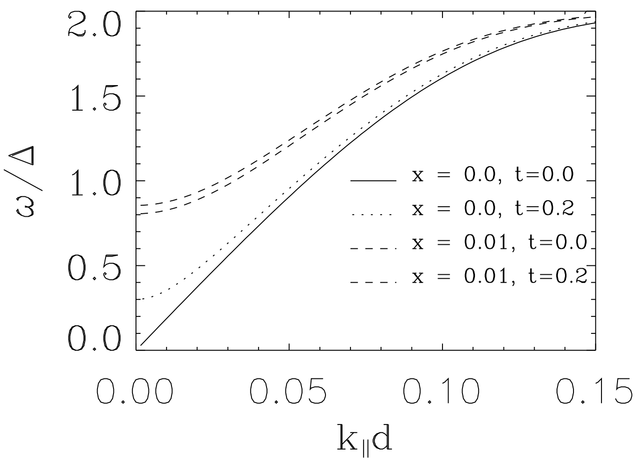

FIG. 3. The dispersion of the out-phase mode (ω_-) in the charged system in the presence of both the single particle tunneling and Josephson tunneling as a function of $k_\parallel$ for a fixed $k_z d = \pi$. Here, $x = T_J/V_0$ and t is the strength of the single particle tunneling with respect to the superconducting energy gap. We use the same parameters as Fig. 2.

$k_z d = \pi$. As emphasized before, the collective mode dispersion is qualitatively independent of the specific tunneling mechanism (i.e., t or T_J), and therefore experiments involving collective modes cannot distinguish between the existing tunneling mechanisms in high-T_c superconductors as has recently been emphasized[6] in a related context.

III. DISCUSSION AND CONCLUSION

We calculate in this paper the collective charge density fluctuation excitation spectra of both the neutral and the charged superconducting bilayerd superlattices with interlayer intra-cell single particle and Josephson tunneling. We use the conserving gauge-invariant ladder diagram approximation in the Nambu-Gorkov formalism. In general, there are two types of density fluctuation modes: in-phase (ω_+) and out-of-phase (ω_-) modes. For neutral superconductors, the out-of-phase collective mode with interlayer tunneling has a plasma gap depending on the tunneling intensity, and the in-phase mode, lying lower in energy, dominates the oscillator strength for all wave vectors. However, for charged superconductors the two phase modes couple to the long range Coulomb interaction differently, and the out-of-phase mode with tunneling dominates the oscillator strength in the long wavelength limit ($k_\parallel \to 0$) and finite k_z. Since we have used two identical 2D layers in each unit cell there is no mode coupling effect in our theory between $\omega_\pm$ modes at the resonant frequency ($\omega_+ \sim \omega_-$). If the two layers forming the unit cell are not identical then there will be a resonant mode coupling effect ("anti-crossing") between the in-phase and the out-of-phase modes around $\omega_+ \approx \omega_-$ resonance point – the nature of this anti-crossing phenomena will be similar to what is seen in the corresponding intrasubband-intersubband collective mode coupling in semiconductor quantum well systems[14]. We have mostly concentrated in the long wavelength regime ($k_\parallel \to 0$) – at large wave vectors there is significant coupling between the collective modes and the pair-breaking excitations, which has been extensively studied in the literature[1,2]. We have also neglected the amplitude fluctuation modes because they usually carry negligible spectral weights compared with the $\omega_\pm$ phase fluctuation modes. We have also used an s-wave ground state symmetry which should be a good approximation[2] even for d-wave cuprate systems as far as the long wavelength collective mode properties are concerned. Our use of a BCS–Fermi liquid model in our theory is more difficult to defend except on empirical grounds[6] and for reasons of simplicity.

Finally, we consider the effect of *intercell* tunneling on the collective mode spectra, which we have so far neglected in our consideration. (Our theory includes both intracell and intercell Coulomb coupling between all the layers, and *intracell* interlayer single electron and Josephson tunneling.) The neglect of intercell tunneling is justified by the fact that $d \gg c$ (e.g., in YBCO $d = 12\mathring{A}$, $c = 3\mathring{A}$). The general effect of intercell tunneling becomes quite complicated theoretically because one has far too many interlayer coupling terms in the Hamiltonian in the presence of *both* intracell and intercell interlayer tunneling involving *both* single particle and Josephson tunneling. It is clear, however, that the main effect of a weak intercell interlayer tunneling (either single particle or Josephson type, or both) would be to cause a 2D to 3D transition in the plasma mode by opening up a small gap in both $\omega_\pm$ modes at long wavelengths (in the charged system). The size of this gap (which is the effective 3D plasma frequency of the k_z-motion of the system) will depend on the intercell tunneling strength. This small gap is the 3D c-axis plasma frequency of the system, which has been the subject of several recent studies in the literature[6,12,15].

The introduction of a weak *intercell* interlayer tunneling will therefore modify our calculated results simply through a shift of the energy/frequency origin in our calculated dispersion curves. The origin of the ordinate (i.e., the energy/frequency axis) in our results will shift from zero to ω_c, where ω_c is the c-axis plasma frequency arising from the *intercell* interlayer hopping. For an effective single band tight binding intercell hopping parameter t_c (i.e., the single electron effective bandwidth in the c-direction is $2t_c$), one obtains $\omega_c = \omega_p t_c d/v_F$, where $\omega_p = [4\pi n_B e^2/(\kappa m)]^{1/2}$ is the effective 3D plasma frequency with the 2D a-b plane band mass m [see Eq. (25)] and v_F is the Fermi velocity in the a-b plane. Note that $\omega_c \ll \omega_p$ because t_c is very small by virtue of weak intercell coupling. Note also that if one de-

fines an effective "3D" c-axis plasma frequency $\omega_{pc} = [4\pi n_B e^2/(\kappa m_c)]^{1/2}$ in analogy with ω_p, where m_c is now the effective mass for electron dynamics along the c-axis, then $\omega_c = \omega_{pc}[t/(2E_F)]^{1/2}$ due to the tight bind nature of c-motion. We emphasize that in the presence of intercell hopping ω_c sets the scale for the lowest energy that a collective mode can have in the multilayer superconductor – ω_c is sometimes referred[3,7,8] to as a Josephson plasmon[12] in the literature. In general, it is difficult to theoretically estimate ω_c in high-T_c materials[6] because the effective t_c (and other parameters) may not be known. It is therefore important to emphasize[6,12] that ω_c can be measured directly from the c-axis plasma edge in reflectivity experiments, (we emphasize that a-b plane plasma edge gives ω_p and the c-axis plasma edge gives ω_c[15]), and such measurements[15] show that ω_c is below the superconducting gap in many high-T_c materials[6]. This implies that the effective c-axis hopping, t_c, in high-T_c materials (either due to single particle hopping or due to Josephson coupling arising from coherent Cooper pair hopping) has to be very small (much smaller than that given by direct band structure calculations) in these systems for the Josephson plasma frequency ω_c to be below the superconducting gap, a point first emphasized by Anderson[12].

The collective mode situation in a bilayer system in the presence of both intracell and intercell interlayer coupling is obviously quite complex, and as emphasized in ref. 12, there could in general be several collective phase fluctuation modes depending on the detailed nature of intracell and intercell interlayer hopping matrix. In the most general bilayer system intercell coupling will give rise to two separate ω_+ plasma bands arising from the two distinct possible intercell interlayer coupling — the two ω_+ bands lying in energy lower that the two $\omega-$ bands in the charged system as we show in this paper. In the most general situation[12], there could be two low energy Josephson plasma frequencies ω_{c1}, ω_{c2} ($>\omega_{c1}$), corresponding to the bottoms of the two ω_+ bands, arising respectively from the larger and the smaller of the intercell interlayer hopping amplitudes. To make things really complicated one of these modes (ω_{c1}) could be below the gap and the other (ω_{c2}) above the gap, (or, both could be below or above the gap). While each of these scenarios is possible, c-axis optical response experimental results on inter-bilayer charge dynamics in $YBCO$ have been interpreted[16] to exhibit only one c-axis plasma edge in the superconducting state with the frequency ω_c between 60 cm^{-1} and 200 cm^{-1}, depending on the oxygen content. There are three possibilities: (1) The two plasma modes ($\omega_{c1} \approx \omega_{c2} \approx \omega_c$) are almost degenerate because the corresponding intercell hopping amplitudes are close in magnitudes; (2) ω_{c2} is much lager than ω_{c1} ($\ll \omega_{c2}$) because the two intercell hopping amplitude are very different in magnitudes (we consider this to be an unlikely scenario); (3) one of the two modes carries very little optical spectral weight and is not showing up in c-axis reflectivity measurements, leaving only the other one as the observed c-axis plasma edge. There is, in principle, a fourth (very unlikely) possibility: the observed plasma edge is really ω_{c2}, and the other mode ω_{c1} ($\ll \omega_{c2}$) is too low in energy to show up in c-axis reflectivity measurements.

Within a *nearest-neighbor* c-axis interlayer coupling model, there is only a *single* intercell hopping amplitude, giving rise to only a single c-axis plasma edge ω_c, which now defines the lowest value that the in-phase collective mode ω_+ can have, $\omega_c \equiv \omega_{c+} \equiv \omega_+(k=0)$ — ω_c is shifted up from zero at long wavelengths due to finite c-axis intercell hopping. The out-of-phase plasma edge, $\omega_{c-} \equiv \omega_-(k=0)$, will obviously lie much higher in energy than $\omega_{c+} \equiv \omega_c$ because the intracell interlayer hopping is much stronger than the intercell interlayer hopping. In particular, even though the ω_{c+} mode may lie in the superconducting gap[16,12], we expect ω_{c-} to lie much above the superconducting gap energy in $YBCO$. A crude qualitative estimate can be made by assuming that the intra- and intercell hopping amplitudes scale as inverse squares of lattice parameters: $t_{\rm intra}/t_{\rm inter} \approx (d/c)^2 = 16$. This then leads to the approximate formula $\omega_{c-} \approx 16^2\ \omega_{c+} = 256\ \omega_c$, which, for $YBCO$, implies that the long wavelength out-of-phase mode should lie between 2 eV and 6 eV, depeding on the oxygen content (assuming that the c-axis plasma edge varies between 60 cm^{-1} and 200 cm^{-1}, as reported in ref. 16, depending on the oxygen content). While there is some minor observable structure in optical experiments at high energies, we cannot find any compelling evidence in favor of the existence of a high energy out-of-phase mode in the currently available experimental data. We feel that a spectroscopic experiment, using, for example, the inelastic electron energy loss spectroscopy which could probe the mode dispersion (and which has been highly successful in studying bulk plasmons in metal films) of the ω_- mode at high energy, may be required to unambiguously observe the out-of-phase collective mode. What we have shown in this paper is that under suitable conditions (finite k and k_z) the ω_- out-of-phase mode carries reasonable spectral weight and should be observable in principle — actual observation, however, awaits experimental investigations using external probes which can study mode dispersion at finite wave vectors (which optical experiments by definition cannot do; they are long wavelength probes).

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